## Supporting Information

## Thermodynamically Self-organized Hole Transport Layer for High-Efficiency Inverted-Planar Perovskite Solar Cells

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(a)

(c)

(b)

(d)


Fig. S1. $J-V$ curves measured by forward (from $\mathrm{J}_{\mathrm{SC}}$ to $\mathrm{V}_{\mathrm{OC}}$, red) and reverse scans (from $\mathrm{V}_{\mathrm{OC}}$ to $\mathrm{J}_{\mathrm{SC}}$, black) of the IP-PSCs containing various PEDOT:PSS films depending on the PSS/PEDOT ratio: (a) PEDOT1:PSS0.5, (b) PEDOT1:PSS2.5, (c) PEDOT1:PSS5.0 and (d) PEDOT1:PSS12.



Fig. S2. (a) Photocurrent density and (b) power conversion efficiency as a function of time for the IP-PSC devices containing PEDOT:PSS films with controlled weight ratios. Data were obtained at the maximum voltage, $\mathrm{Vmax}=0.63$ (PEDOT1:PSS0.5), 0.77 (PEDOT1:PSS2.5), 0.8 (PEDOT1:PSS5.0) and 0.76 (PEDOT1:PSS12), without pre-exposure under 1 sun illumination.


Fig. S3. (a) Schematic of a hole-transport process from perovskite layer to ITO electrode. (b) Schematic of a hole-transport process from perovskite layer to ITO electrode via PEDOT1:PSS5.0 layer.


Fig. S4. Top-view SEM image of $\mathrm{MAPbI}_{3}$ films on the various PEDOT:PSS layers depending on the PSS/PEDOT ratio: (a-b) PEDOT1:PSS0.5, (c-d) PEDOT1:PSS2.5, (e-f) PEDOT1:PSS5.0 and (g-h) PEDOT1:PSS12. Scale bar, $1000 \mathrm{~nm}(\mathrm{a}, \mathrm{c}, \mathrm{e}, \mathrm{g})$ and $500 \mathrm{~nm}(\mathrm{~b}, \mathrm{~d}$, f, h).


Fig. S5. Cross-sectional SEM images of the $\mathrm{MAPbI}_{3}$ films on the various PEDOT:PSS layers depending on the PSS/PEDOT ratio: (a) PEDOT1:PSS0.5, (b) PEDOT1:PSS2.5, (c) PEDOT1:PSS5.0 and (d) PEDOT1:PSS12.


Fig. S6. XRD patterns of $\mathrm{MAPbI}_{3}$ films on the various PEDOT:PSS layers depending on the PSS/PEDOT ratio


Fig. S7. Normalized EQE spectra of the IP-PSCs containing various PEDOT:PSS layers depending on the PSS/PEDOT ratio.

